Quantum-Mechanical Interference Between Optical Transitions:
The Effect of Laser Intensity Noise

15 May 1999

Prepared by

J. C. CAMPARO
Electronics Technology Center
Technology Operations
The Aerospace Corporation

and

P. LAMBROPOULOS
Max-Planck Institute fur Quantenoptik
Germany

19990603 163

Engineering and Technology Group

**THE AEROSPACE
CORPORATION**
El Segundo, California
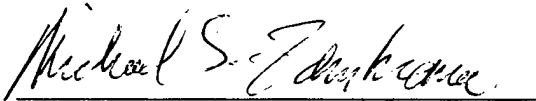
DTIC QUALITY INSPECTED 4

Michael Zambrana
SMC/AXE

REPORT DOCUMENTATION PAGE

1. AGENCY USE ONLY (*Leave blank*)	2. REPORT DATE 15 May 1999	3. REPORT TYPE AND DATES COVERED

4. TITLE AND SUBTITLE Quantum-Mechanical Interference Between Optical Transitions: The Effect of Laser Intensity Noise	5. FUNDING NUMBERS F04701-93-C-0094
6. AUTHOR(S) Camparo, J. C., The Aerospace Corporation; and Lambropoulos, P., Max-Planck Institute fur Quantenoptik	

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) The Aerospace Corporation Technology Operations El Segundo, CA 90245-4691	8. PERFORMING ORGANIZATION REPORT NUMBER TR-98(8555)-9

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) Space and Missile Systems Center Air Force Materiel Command 2430 E. El Segundo Blvd. Los Angeles Air Force Base, CA 90245	10. SPONSORING/MONITORING AGENCY REPORT NUMBER SMC-TR-99-18

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited	12b. DISTRIBUTION CODE

13. ABSTRACT (*Maximum 200 words*)

In a previous publication [Phys. Rev. A **55**, 552 (1997)], we considered the effect of laser phase fluctuations on three-photon–one-photon phase control of resonance-enhanced photoionization. Here we extend those studies by considering the role of laser intensity noise in addition to laser phase noise. While our results indicate that relative intensity fluctuations between the fundamental field and its third harmonic have a significant effect on control, the contrast between constructive and destructive interference is nonetheless two orders of magnitude, even under (reasonable) worst case situations. Consequently, neither laser intensity nor phase fluctuations appear to pose a serious impediment to the efficient phase control of atomic and molecular processes.

14. SUBJECT TERMS Atomic clocks Spectroscopy Optical pumping	15. NUMBER OF PAGES 7
	16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT

Quantum-mechanical interference between optical transitions: The effect of laser intensity noise

J. C. Camparo[1] and P. Lambropoulos[2,3]

[1]*Electronics Technology Center, The Aerospace Corporation, P.O. Box 92957, Los Angeles, California 90009*

[2]*Max-Planck Institute für Quantenoptik, D-1046 Garching, Germany*

[3]*FORTH Institute of Electronic Structure and Laser, P.O. Box 1527, Heraklion 71110, Crete, Greece*

and Department of Physics, University of Crete, Crete, Greece

In a previous publication [Phys. Rev. A **55**, 552 (1997)] we considered the effect of laser phase fluctuations on three-photon–one-photon phase control of resonance-enhanced photoionization. Here we extend those studies by considering the role of laser intensity noise in addition to laser phase noise. While our results indicate that relative intensity fluctuations between the fundamental field and its third harmonic have a significant effect on control, the contrast between constructive and destructive interference is nonetheless two orders of magnitude, even under (reasonable) worst case situations. Consequently, neither laser intensity nor phase fluctuations appear to pose a serious impediment to the efficient phase control of atomic and molecular processes. [S1050-2947(99)00403-5]

In $(3+1)$-photon phase control, an optical transition is excited via a three-photon and a one-photon pathway as illustrated in Fig. 1 for the case of Xe multiphoton ionization [1,2]. The field at the fundamental frequency ω_1 has a phase ϕ_1, while the third-harmonic field with frequency ω_3 has a phase ϕ_3. (Following common practice, the subscripts indicate the harmonic nature of the field and not the number of photons required for the transition.) These phases are controlled by passing the two fields through a dispersive medium of length L with indices of refraction n_1 and n_3 for the fundamental and third-harmonic fields, respectively. The rate of excitation is proportional to the square of the total transition amplitude and hence proportional to $\Omega_1^2[1+f^2+2f\cos(\theta)]$, where Ω_1 is the fundamental field's three-photon Rabi frequency, f is the ratio of the one-photon to three-photon Rabi frequency (i.e., $f\equiv\Omega_3/\Omega_1$), and the relative phase $\theta=\phi_3-3\phi_1$. (For the purposes of our work here, we have ignored any constant phase difference between the two quantum-mechanical paths since even in those cases where it might be nonzero it can be incorporated into the relative phase difference between the two fields.) As the relative phase difference between the two fields is varied, typically by changing the vapor pressure of the dispersive medium, the rate of excitation exhibits constructive and destructive interference. The maximum contrast between constructive and destructive interference occurs when f equals unity.

In a previous publication [3], we considered the influence of laser phase noise on this control process. Since the dispersive medium's propagation constant depends on frequency, the relative phase between the two fields is a stochastic quantity by virtue of the stochastic nature of the laser frequency: $\theta(t)=\theta_0+\delta\theta(t)$, where

$$\theta_0=\frac{3\bar{\omega}_1 L}{c}[n_3-n_1], \tag{1a}$$

$$\delta\theta(t)=\frac{3\,\delta\omega_1(t)L}{c}[n_3-n_1]. \tag{1b}$$

Here $\bar{\omega}_1$ and $\delta\omega_1(t)$ refer to the mean frequency and stochastic frequency fluctuation of the fundamental field, respectively. Somewhat surprisingly, our results indicated that orders of magnitude of contrast could be maintained even when employing phase diffusion fields (PDFs) with a linewidth of 3 cm^{-1}. Here we extend our previous investigation by considering fields with amplitude noise as well as phase noise.

One expects an amplitude noisy field to influence $(3+1)$-photon phase control since f (in addition to θ) may now be a stochastic quantity. Unfortunately, little detailed information is available regarding the relationship between a fundamental field's stochastic characteristics and those of a harmonic field generated within a nonlinear medium, specifically in the sense of coordinated experimental and theoretical studies mapping the stochastic characteristics of the fundamental field to the harmonic field. Of course, on general grounds it is known that a third-order process induced
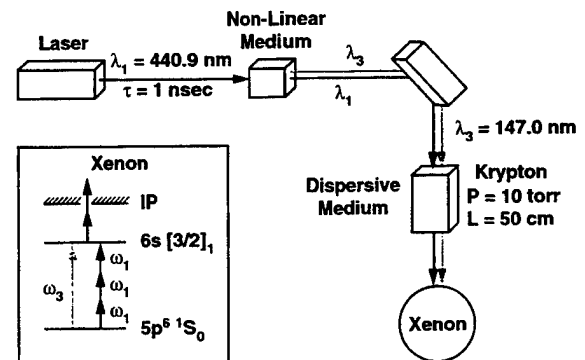
FIG. 1. Standard $(3+1)$-photon phase-control experiment. A fundamental field from a laser is tripled in a nonlinear medium and then both the fundamental and third harmonic pass through a dispersive medium. Since the refractive indices of the dispersive medium for the fundamental and third-harmonic fields are different, a relative phase difference between the two fields is created. Our numerical simulations consider $(3+1)$-photon phase control of xenon photoionization, where IP stands for ionization potential.

by a chaotic field should depend on a third-order intensity correlation function, which involves factors of 3!. However, the 3! term appearing in the third-harmonic field intensity should be similar to a 3! term appearing in the three-photon Rabi frequency, implying that f will nonetheless be constant, a conclusion borne out by results to be presented below.

To go beyond these general considerations, consider the nonlinear polarization equation, where the nonlinear medium's third-harmonic polarization P_3 generates the harmonic field E_3. Defining χ_3 as the nonlinear electric susceptibility, we have [4]

$$P_3(t)=\chi_3(\omega_1)\left(\frac{|E_1|}{2}\right)^3 \exp[-i3\omega_1 t]+\text{c.c.}, \quad (2a)$$

where [5]

$$\chi_3(\omega_1)\approx\sum_{n,m,q}\frac{\langle g|r|q\rangle\langle q|r|m\rangle\langle m|r|n\rangle\langle n|r|g\rangle}{(\omega_{qg}-3\omega_1)(\omega_{mg}-2\omega_1)(\omega_{ng}-\omega_1)}, \quad (2b)$$

and $\Omega_3 \sim \chi_3(\omega_1)$. In Eq. (2b), $|g\rangle$ is the ground state and $\omega_{ij}=(E_i-E_j)/\hbar$. Given the stochastic nature of ω_1, each of the resonance factors in the denominator of Eq. (2b) will fluctuate and this will give rise to fluctuations in χ_3 and hence the one-photon Rabi frequency. Typically, however, the fluctuations of χ_3 are dominated by only one of the factors, specifically the two-photon resonance term. Further, since the fundamental field's frequency fluctuations will likely be small in comparison to the resonance detuning, Eq. (2b) can be rewritten as

$$\chi_3(\omega_1)\approx\sum_{n,m,q}\frac{\langle g|r|q\rangle\langle q|r|m\rangle\langle m|r|n\rangle\langle n|r|g\rangle}{(\omega_{qg}-3\bar{\omega}_1)(\omega_{mg}-2\bar{\omega}_1)(\omega_{ng}-\bar{\omega}_1)}$$
$$\times\left[1+\frac{2\delta\omega_1}{(\omega_{mg}-2\bar{\omega}_1)}\right]. \quad (3)$$

(Notice that the correlation between χ_3 and the laser frequency fluctuations can be either positive or negative, depending on the detuning of the laser from the two-photon resonance.) Writing χ_3 in terms of its average value $\bar{\chi}_3$ and a mean-zero stochastic quantity $\delta\chi_3(t)$, Eqs. (2a) and (3) indicate that f is subject to additive noise and that under optimal conditions (i.e., $\langle f\rangle=1$)

$$f(t)=\left(1+\frac{\delta\chi_3(t)}{\bar{\chi}_3}\right)=\left(1+\frac{2\delta\omega_1(t)}{\Delta_2}\right), \quad (4)$$

where Δ_2 is the two-photon detuning in the nonlinear medium. Thus the stochastic variations of both θ and f in (3+1)-photon phase control derive from the fundamental field's frequency fluctuations.

The fundamental field is described in terms of our standard model for stochastic laser characteristics [6]: $E_1(t)=E_0(1+\epsilon)\cos[(\bar{\omega}_1+\delta\omega_1)t]$, where

$$\langle\epsilon(t)\epsilon(t+\tau)\rangle=\frac{\gamma}{\omega_f}\exp[-\omega_f|\tau|] \quad (5a)$$
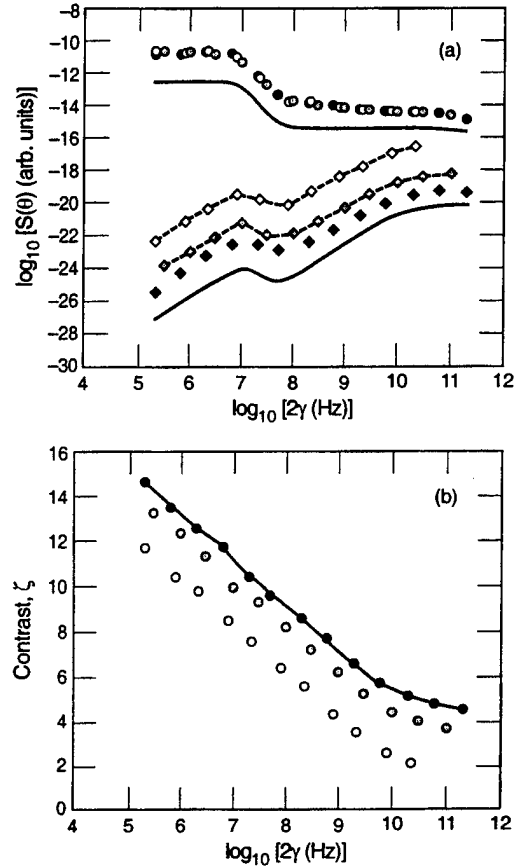
and

FIG. 2. (a) Signal amplitude versus laser linewidth. Circles correspond to constructive interference, while diamonds correspond to destructive interference: Filled black symbols correspond to $\Delta_2 = \infty$, gray symbols correspond to $\Delta_2 = 100\,\text{cm}^{-1}$, and open symbols correspond to $\Delta_2 = 10\,\text{cm}^{-1}$. The solid lines correspond to previous results with a PDF, while the dashed lines are simply meant as an aid to guide the eye. (b) Contrast ζ versus laser linewidth. Symbol shading is as in (a) and again the solid line corresponds to previous PDF results.

$$\langle\delta\omega(t)\delta\omega(t+\tau)\rangle=\gamma\beta\exp[-\beta|\tau|]. \quad (5b)$$

Basically, γ defines the linewidth of the nearly Lorentzian line shape, β is a cutoff parameter for the line-shape wings, and ω_f is a bandwidth parameter associated with the fundamental field's amplitude fluctuations.

Our computational procedure is equivalent to that discussed previously [3]. We generate a realization of the fundamental field's frequency and amplitude fluctuations and then numerically integrate the relevant Xe density matrix equations for a 1-ns Gaussian pulse using a Runge-Kutta-Fehlberg technique [7], including now the third-harmonic field's additive noise. The signal that exhibits phase control is the total ionization $S(\theta_0)$ produced by the field during the pulse. Computing two ionization signals $S(0)$ and $S(\pi)$, we define the contrast of phase control as $\zeta = \log_{10}[S(0)/S(\pi)]$. For the results to be reported here, $\omega_f = 3\gamma$ and $\beta = 100\gamma$, indicating that the fundamental field was a near-chaotic field with an essentially Lorentzian line shape. The peak intensity of the fundamental field was $10^8\,\text{W/cm}^2$ (i.e., weak-field conditions) and the transition ac Stark shift was set to zero.

Figure 2(a) shows the logarithm of the signal amplitude as a function of the fundamental field's linewidth parameter (i.e., 2γ). Circles correspond to constructive interference signals, while diamonds correspond to destructive interference signals: For the black symbols $\Delta_2 = \infty$ (i.e., no fluctuations in f), for the gray symbols $\Delta_2 = 100\,\mathrm{cm}^{-1}$, and for the open symbols $\Delta_2 = 10\,\mathrm{cm}^{-1}$. The solid lines correspond to our previous results with a PDF, while the dashed lines are simply meant as an aid to guide the eye. Considering for the moment the case of $\Delta_2 = \infty$, laser intensity noise increases the signal amplitude dramatically for both constructive and destructive interference, as one might expect. Specifically, the signal enhancement factor is on the order of 50–70. (Note that for a nonresonant five-photon ionization process induced by a chaotic field, one would expect a 5! signal enhancement [8].) As additive noise for the harmonic field is increased, there is little change in the constructive interference signal amplitude, but a noticeable increase in the destructive interference signal. For example, in the case of $\Delta_2 = 10\,\mathrm{cm}^{-1}$ at $2\gamma = 1$ MHz, the additive noise increases the destructive signal amplitude by more than three orders of magnitude even though $\delta f_{\mathrm{rms}} \sim 3 \times 10^{-5}$. This behavior is reflected in the contrast of phase control shown in Fig. 2(b),

where the shading of the symbols is the same as in Fig. 2(a) and the solid line again corresponds to our previous results with a PDF.

Clearly, Fig. 2(b) demonstrates that a small amount of additive noise can have a significant influence on the contrast of phase control. However, even in the worst (reasonable) case of $\Delta_2 = 10\,\mathrm{cm}^{-1}$ and a field linewidth of approximately $1\,\mathrm{cm}^{-1}$, the phase control contrast is two orders of magnitude. Thus, in an absolute sense phase control appears to be extremely tolerant of stochastic fields. The absolute degree of control predicted here, even in the worst case, is much better than what has yet been demonstrated experimentally, and from this we conclude that $(3+1)$-photon phase control is limited at present by processes other than those discussed here. For example, as noted by Chen and Elliot [9], overlap and focusing of the fundamental field and its first harmonic are extremely important for optimum phase control contrast. Given the present results, it is quite likely that these other experimental issues are the major processes limiting "orders of magnitude" $(3+1)$-photon phase control and not any inherent stochastic fluctuations of the laser field.

[1] M. Shapiro, J. W. Hepburn, and P. Brumer, Chem. Phys. Lett. **149**, 451 (1988); P. Brumer and M. Shapiro, Acc. Chem. Res. **22**, 407 (1989); C. K. Chan, P. Brumer, and M. Shapiro, J. Chem. Phys. **94**, 2688 (1991).

[2] We note that the $(3+1)$-photon mechanism had been proposed previously as an explanation for the suppression of three-photon excitation in dense atomic vapors. See, for example, D. J. Jackson and J. J. Wynne, Phys. Rev. Lett. **49**, 543 (1982).

[3] J. C. Camparo and P. Lambropoulos, Phys. Rev. A **55**, 552 (1997).

[4] Equation (2) is a generalization of Mathur, Tang, and Happer's relationship between induced polarization and the electric field: B. S. Mathur, H. Y. Tang, and W. Happer, Phys. Rev. A **2**, 648 (1970).

[5] J. F. Reintjes, *Nonlinear Optical Parametric Processes in Liquids and Gases* (Academic, Orlando, 1984).

[6] J. C. Camparo and P. Lambropoulos, Phys. Rev. A **47**, 480 (1993).

[7] W. Cheney and D. Kincaid, *Numerical Mathematics and Computing* (Brooks Cole, Monterey, CA, 1985); W. H. Press and S. A. Teukolsky, Comput. Phys. **6**, 188 (1992).

[8] A. T. Georges and P. Lambropoulos, in *Advances in Electronics and Electron Physics* (Academic, New York, 1980), Vol. 54.

[9] C. Chen and D. S. Elliot, Phys. Rev. Lett. **65**, 1737 (1990).

TECHNOLOGY OPERATIONS

The Aerospace Corporation functions as an "architect-engineer" for national security programs, specializing in advanced military space systems. The Corporation's Technology Operations supports the effective and timely development and operation of national security systems through scientific research and the application of advanced technology. Vital to the success of the Corporation is the technical staff's wide-ranging expertise and its ability to stay abreast of new technological developments and program support issues associated with rapidly evolving space systems. Contributing capabilities are provided by these individual Technology Centers:

Electronics Technology Center: Microelectronics, VLSI reliability, failure analysis, solid-state device physics, compound semiconductors, radiation effects, infrared and CCD detector devices, Micro-Electro-Mechanical Systems (MEMS), and data storage and display technologies; lasers and electro-optics, solid state laser design, micro-optics, optical communications, and fiber optic sensors; atomic frequency standards, applied laser spectroscopy, laser chemistry, atmospheric propagation and beam control, LIDAR/LADAR remote sensing; solar cell and array testing and evaluation, battery electrochemistry, battery testing and evaluation.

Mechanics and Materials Technology Center: Evaluation and characterization of new materials: metals, alloys, ceramics, polymers and composites; development and analysis of advanced materials processing and deposition techniques; nondestructive evaluation, component failure analysis and reliability; fracture mechanics and stress corrosion; analysis and evaluation of materials at cryogenic and elevated temperatures; launch vehicle fluid mechanics, heat transfer and flight dynamics; aerothermodynamics; chemical and electric propulsion; environmental chemistry; combustion processes; spacecraft structural mechanics, space environment effects on materials, hardening and vulnerability assessment; contamination, thermal and structural control; lubrication and surface phenomena; microengineering technology and microinstrument development.

Space and Environment Technology Center: Magnetospheric, auroral and cosmic ray physics, wave-particle interactions, magnetospheric plasma waves; atmospheric and ionospheric physics, density and composition of the upper atmosphere, remote sensing, hyperspectral imagery; solar physics, infrared astronomy, infrared signature analysis; effects of solar activity, magnetic storms and nuclear explosions on the earth's atmosphere, ionosphere and magnetosphere; effects of electromagnetic and particulate radiations on space systems; component testing, space instrumentation; environmental monitoring, trace detection; atmospheric chemical reactions, atmospheric optics, light scattering, state-specific chemical reactions and radiative signatures of missile plumes, and sensor out-of-field-of-view rejection.